

FORM PTO-1390 (REV. 11-2000)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER
				0992-0127P
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (If known, see 37 CFR 1.5) <b>10/06901</b>
INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE		PRIORITY DATE CLAIMED	
PCT/JP00/05737	August 25, 2000		August 27, 1999	
<b>TITLE OF INVENTION</b> LAMINATE, PROCESS FOR PRODUCING IT AND USE THEREOF				
<b>APPLICANT(S) FOR DO/EO/US</b> SAWADA, Yuji and NAKASHIMA, Yasushi				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
<ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</li> <li><input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).</li> <li><input checked="" type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).</li> <li><input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))             <ol style="list-style-type: none"> <li><input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li><input checked="" type="checkbox"/> has been transmitted by the International Bureau. (WO 01/17043)</li> <li><input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).</li> </ol> </li> <li><input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2))             <ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> is transmitted herewith.</li> <li><input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4)</li> </ol> </li> <li><input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))             <ol style="list-style-type: none"> <li><input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li><input type="checkbox"/> have been transmitted by the International Bureau.</li> <li><input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li><input checked="" type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li><input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).</li> <li><input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).</li> <li><input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).</li> </ol>				
Items 11. to 20. below concern document(s) or information included:				
<ol style="list-style-type: none"> <li><input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98, Form PTO-1449(s), and International Search Report (PCT/ISA/210) with 3 cited document(s).</li> <li><input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li><input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment.</li> <li><input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> <li><input type="checkbox"/> A substitute specification.</li> <li><input type="checkbox"/> A change of power of attorney and/or address letter.</li> <li><input type="checkbox"/> A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821-1.825.</li> <li><input type="checkbox"/> A second copy of the published international application under 35 U.S.C. 154(d)(4).</li> <li><input type="checkbox"/> A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).</li> <li><input checked="" type="checkbox"/> Other items or information:             <ol style="list-style-type: none"> <li>1.) International Preliminary Examination Report (PCT/IPEA/409)</li> <li>2.) Form PCT/IB/304</li> <li>3.) Form PCT/IB/308</li> <li>4.) Zero (0) Sheets of Formal Drawings</li> </ol> </li> </ol>				

U.S. APPLICATION NO (if known, see 37 CFR 1.5) <b>10/ NEW 069001</b>	INTERNATIONAL APPLICATION NO <b>PCT/JP00/05737</b>	ATTORNEY'S DOCKET NUMBER <b>0992-0127P</b>																																																				
<p><input checked="" type="checkbox"/> The following fees are submitted:</p> <p><b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5):</b></p> <p>Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO. .... <b>\$1,040.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$890.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO. .... <b>\$740.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$710.00</b></p> <p>International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4). .... <b>\$100.00</b></p> <p><b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b></p> <p><b>\$ 890.00</b></p> <p><b>Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).</b> <b>\$ 0.00</b></p> <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 25%;">CLAIMS</th> <th style="width: 25%;">NUMBER FILED</th> <th style="width: 25%;">NUMBER EXTRA</th> <th style="width: 25%;">RATE</th> </tr> </thead> <tbody> <tr> <td>Total Claims</td> <td>24 - 20 =</td> <td>4</td> <td>X \$18.00 \$ 72.00</td> </tr> <tr> <td>Independent Claims</td> <td>10 - 3 =</td> <td>7</td> <td>X \$84.00 \$ 588.00</td> </tr> <tr> <td colspan="2">MULTIPLE DEPENDENT CLAIM(S) (if applicable)</td> <td>None</td> <td>+ \$280.00 \$ 0.00</td> </tr> <tr> <td colspan="4" style="text-align: center;"><b>TOTAL OF ABOVE CALCULATIONS = \$ 1,550.00</b></td> </tr> <tr> <td colspan="4"><input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2. <b>\$ 0.00</b></td> </tr> <tr> <td colspan="4" style="text-align: center;"><b>SUBTOTAL = \$ 1,550.00</b></td> </tr> <tr> <td colspan="2">Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).</td> <td style="text-align: center;">+</td> <td style="text-align: center;">\$ 0.00</td> </tr> <tr> <td colspan="4" style="text-align: center;"><b>TOTAL NATIONAL FEE = \$ 1,550.00</b></td> </tr> <tr> <td colspan="2">Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property</td> <td style="text-align: center;">+</td> <td style="text-align: center;">\$ 40.00</td> </tr> <tr> <td colspan="4" style="text-align: center;"><b>TOTAL FEES ENCLOSED = \$ 1,590.00</b></td> </tr> <tr> <td colspan="2"></td> <td style="text-align: center;">Amount to be: refunded</td> <td style="text-align: center;">\$</td> </tr> <tr> <td colspan="2"></td> <td style="text-align: center;">charged</td> <td style="text-align: center;">\$</td> </tr> </tbody> </table> <p>a. <input checked="" type="checkbox"/> A check in the amount of <u>\$ 1,590.00</u> to cover the above fees is enclosed.</p> <p>b. <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>02-2448</u>.</p> <p><b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b></p> <p>Send all correspondence to:  <b>Birch, Stewart, Kolasch &amp; Birch, LLP or Customer No. 2292</b>  <b>P.O. Box 747</b>  <b>Falls Church, VA 22040-0747</b>  <b>(703) 205-8000</b></p> <p>Date: <u>February 20, 2002</u></p> <p style="text-align: right;">By   <b>Marc S. Weiner, #32,181</b></p>		CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	Total Claims	24 - 20 =	4	X \$18.00 \$ 72.00	Independent Claims	10 - 3 =	7	X \$84.00 \$ 588.00	MULTIPLE DEPENDENT CLAIM(S) (if applicable)		None	+ \$280.00 \$ 0.00	<b>TOTAL OF ABOVE CALCULATIONS = \$ 1,550.00</b>				<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2. <b>\$ 0.00</b>				<b>SUBTOTAL = \$ 1,550.00</b>				Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		+	\$ 0.00	<b>TOTAL NATIONAL FEE = \$ 1,550.00</b>				Fee for recording the enclosed assignment (37 CFR 1.21(h)). 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PATENT  
0992-0127P

IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: SAWADA, Yuji et al.

Int'l. Appl. No.: PCT/JP00/05737

Appl. No.: New Group:

Filed: February 20, 2002 Examiner:

For: LAMINATE, PROCESS FOR PRODUCING IT  
AND USE THEREOF

PRELIMINARY AMENDMENT

**BOX PATENT APPLICATION**

Assistant Commissioner for Patents  
Washington, DC 20231

February 20, 2002

Sir:

The following Preliminary Amendments and Remarks are respectfully submitted in connection with the above-identified application.

AMENDMENTS

IN THE SPECIFICATION:

Please amend the specification as follows:

Before line 1, insert --This application is the national phase under 35 U.S.C. § 371 of PCT International Application No. PCT/JP00/05737 which has an International filing date of August 25, 2000, which designated the United States of America.--

IN THE CLAIMS:

Please amend the claims as follows:

5. (Amended) The laminate as claimed in claim 1, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

6. (Amended) The laminated as claimed in claim 1, wherein the surface-treated layer is constituted of a chemically treated layer.

7. (Amended) The laminated as claimed in claim 1, wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

8. (Amended) The laminated as claimed in claim 1, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

14. (Amended) A seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in claim 1.

15. (Amended) A battery comprising a seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in claim 1.

16. (Amended) A seal film or sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in claim 3.

17. (Amended) A secondary battery comprising a seal film for sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in claim 3.

20. (Amended) The flexible package as claimed in claim 18, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

21. (Amended) The flexible package as claimed in claim 18, wherein the surface-treated layer is constituted of a chemically treated layer.

22. (Amended) The flexible package as claimed in claim 18, wherein the primer coating layer is that of thermosetting type

based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

23. (Amended) The flexible package as claimed in claim 18, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

REMARKS

The specification has been amended to provide a cross-reference to the previously filed International Application.

The claims have been amended to remove multiple dependencies.

Entry of the above amendments is earnestly solicited. An early and favorable first action on the merits is earnestly solicited.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. § 1.16 or under 37 C.F.R. § 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By   
Marc S. Weiner, #32,181

MSW/rem  
0992-0127P

P.O. Box 747  
Falls Church, VA 22040-0747  
(703) 205-8000

Attachment: VERSION WITH MARKINGS TO SHOW CHANGES MADE

VERSION WITH MARKINGS TO SHOW CHANGES MADE

The specification has been amended to provide a cross-reference to the previously filed International Application.

IN THE CLAIMS:

The claims have been amended as follows:

5. (Amended) The laminate as claimed in [any one of claims 1 to 4] claim 1, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.
6. (Amended) The laminated as claimed in [any one of claims 1 to 5] claim 1, wherein the surface-treated layer is constituted of a chemically treated layer.
7. (Amended) The laminated as claimed in [any one of claims 1 to 6] claim 1, wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.
8. (Amended) The laminated as claimed in [any one of claims 1 to 7] claim 1, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

14. (Amended) A seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in [any one of claims 1, 2 and 5 to 9] claim 1.

15. (Amended) A battery comprising a seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in [any one of claims 1, 2 and 5 to 9] claim 1.

16. (Amended) A seal film or sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in [any one of claims 3 to 9] claim 3.

17. (Amended) A secondary battery comprising a seal film for sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in [any one of claims 3 to 9] claim 3.

20. (Amended) The flexible package as claimed in [18 or 19] claim 18, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

21. (Amended) The flexible package as claimed in [any one of claims 18 to 20] claim 18, wherein the surface-treated layer is constituted of a chemically treated layer.

22. (Amended) The flexible package as claimed in [any one of claims 18 to 21] claim 18, wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

23. (Amended) The flexible package as claimed in [any one of claims 18 to 22] claim 18, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

SPECIFICATION

LAMINATE, PROCESS FOR PRODUCING IT  
AND USE THEREOF

FIELD OF THE INVENTION

The present invention relates to a laminate obtained by laminating a metal and an adhesive resin to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, in particular, a laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery; to a process for producing it; and to use thereof.

BACKGROUND OF THE INVENTION

As an enclosure bag for enclosing a positive electrode, a negative electrode, electrolytic solution and so on constituting a non-aqueous electrolyte battery and taking out each of the lead wires for the positive and the negative electrode to the outside and for enclosing also these lead wires, a composite material laminated with a plastic in which a metal foil or a vacuum-deposited metal layer is interposed in a sandwich-like form is used. Such an enclosure bag requires that the plastic held contacting with the electrolytic solution on at least inner side does not

dissolve in the electrolyte, so that an enclosure bag is proposed, in which the sealing reliability is remarkably increased by using a maleic acid-modified polyolefin resin for the inner most layer and by constructing the heat sealing part by this maleic acid-modified polyolefin resin (Japanese Patent Kokai Hei 9-283101 A).

The maleic acid-modified polyolefin resin is superior in the adhesion onto metal and in the heat-sealing performance, so that it is used in general as an adhesive resin. When, however, it is attempted to use the maleic acid-modified resin as a sealing film for batteries, as mentioned above, an interlayer separation may occur after the film is held continued in contact with the electrolytic solution and it cannot be used as a sealing film, though it exhibits superior adhesive strength directly after it is laminated with metal.

A non-aqueous electrolyte is one in which a salt is dissolved in an organic polar solvent. The reason of occurrence of the interlayer separation is assumed to be due to that the organic polar solvent and the salt will penetrate through the maleic acid-modified polyolefin layer and reach the metal layer to cause a reaction with the metal, since the maleic acid-modified polyolefin and the salt have an affinity to the maleic acid-modified polyolefin resin which is polarized by maleic acid. Conventional laminates have a problem that such an interlayer separation cannot be avoided. Further, it is necessary to attach a protective film to

the positive and negative electrodes, since they are metal and subject to a possibility of occurrence of short-circuit. Here also a problem occurs in which the protective film peels off due to the same reason as the sealing film.

The object of the present invention is to provide a laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, in particular, for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, which laminate is superior in the adhesive strength between the metal layer and the adhesive resin layer and in the fastness to organic polar solvents or to salts and which does not suffer from interlayer separation even when being held in contact with a non-aqueous electrolyte or the like, and a process for its efficient production as well as its uses, in order to solve the problems mentioned above.

#### DISCLOSURE OF THE INVENTION

The present invention consists in the following laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, process for its production and its uses:

- (1) A laminate to be served as seal film for sealing the electrolyte of battery or as protection film for

protecting the electrode part of battery (referred to hereinafter sometimes simply as "the laminate"), comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.

(2) A laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery (referred to hereinafter sometimes simply as "the laminate"), comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer,

a layer of a primer coating formed over the said surface-treated layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.

(3) A laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery (referred to hereinafter sometimes simply as "the laminate"), comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.

(4) A laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery (referred to hereinafter sometimes simply as "the laminate"), comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer,

a layer of a primer coating formed over the said surface-treated layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.

(5) The laminate as defined in any one of the above (1) to (4), wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

(6) The laminate as defined in any one of the above (1) to (5), wherein the surface-treated layer is constituted of a chemically treated layer.

(7) The laminate as defined in any one of the above (1) to (6), wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

(8) The laminate as defined in any one of the above (1) to (7), wherein the adhesive resin is a

graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

(9) The laminate as defined in the above (8), wherein the graft-modified polyolefin resin is a graft-modified polyethylene resin or a graft-modified polypropylene resin.

(10) A process for producing a laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising

forming a surface-treated layer over the surface of a metal layer and

laminating on the surface-treated layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

(11) A process for producing a laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising

forming a surface-treated layer over the surface of a metal layer,

forming a primer coating layer over the said surface-treated layer and

laminating on the primer coating layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

(12) A process for producing a laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the

electrode part of secondary battery, comprising

forming a surface-treated layer over the surface of a metal layer and

laminating on the surface-treated layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

(13) A process for producing a laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, comprising

forming a surface-treated layer over the surface of a metal layer,

forming a primer coating layer over the said surface-treated layer and

laminating on the primer coating layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

(14) A seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as defined in any one of the above (1), (2) and (5) to (9).

(15) A battery comprising a seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as defined in any one of the above (1), (2) and (5) to (9).

(16) A seal film for sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is

made from the laminate as defined in any one of the above (3) to (9).

(17) A secondary battery comprising a seal film for sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as defined in any one of the above (3) to (9).

(18) A flexible package made from a laminate comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.

(19) A flexible package made from a laminate comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer,

a layer of a primer coating formed over the said surface-treated layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.

(20) The flexible package as defined in the above (18) or (19), wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

(21) The flexible package as defined in any one of the above (18) to (20), wherein the surface-treated layer is constituted of a chemically treated layer.

(22) The flexible package as defined in any one of the above (18) to (21), wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

(23) The flexible package as defined in any one of the above (18) to (22), wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

(24) The flexible package as defined in the above (23), wherein the graft-modified polyolefin resin is a graft-modified polyethylene resin or a graft-modified polypropylene resin.

In the present invention, foils, sheets, plates and the like, which are used conventionally for laminates with adhesive resin, of, for example, aluminum, nickel, copper, iron and other simple substance metals, can be used.

The surface-treated layer formed on the surface of metal layer is a coating film layer formed by a surface-treatment for forming an inert protective film on a metal surface, for which chemically treated layers treated by an acid, such as chromic acid, phosphoric acid or so on, or an oxide film layer by an anodic oxidation or so on, may be exemplified. Among them, chemical treatment is preferred for the conventional

operation and performance cost.

In the chemical treatment using chromic acid, phosphoric acid or so on, the metal, such as aluminum etc., is immersed in the chemical treatment bath containing chromic acid (salt) containing 0.05 - 5 % by weight of trivalent chromium and/or phosphoric acid (salt) of pH 2 - 4 at 23 - 80 °C for 1 second - 5 minutes to form a chemically treated film containing chromium. Among them, treatment by a chemical treating liquor containing chromic acid (salt) and phosphoric acid (salt) is preferred.

In the case of anodic oxidation, a metal, such as aluminum, is immersed in the electrolytic solution (5 - 25 % by weight of sulfuric acid with 1 - 5 g/l of aluminum sulfate) maintained at 15 - 25 °C and is treated by direct current 15 - 20 V for 1 - 5 minutes to build up a passive film.

For the primer coating layer to be formed on the surface-treated layer, a primer paint used usually for base coat for coating on metal surface is used. For such a primer paint, a thermosetting type primer paint based on, for example, epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane, can be used. Among them, a primer paint based on epoxy is preferred, since it is superior in the adhesion onto a metal layer and onto a layer of an adhesive resin constituted of a polyolefin modified by carboxyl group or by a derivative thereof and in the durability than other primer paints, such as those based on urethane and so on.

For these primer paints, commercial products can be used. As commercial primer paints based on epoxy, DUFLON K300 and ORGAPLASUF 30NC of Nippon Paint Co., Ltd., EPOMATE B002 and EPIKOTE 828 of Yuka Shell K.K. and ETON 2100E of Kawakami Paint Mfg. Co., Ltd. may be exemplified and, as commercial primer paints based on urethane, CORONATE L and NIPPORAN 1100 of Nippon Polyurethane Industry Co., Ltd. and A-310 and A-3 of Takeda Chemical Ind., Ltd. and, as commercial primer paints based on imine, P-1000 of Nippon Shokubai Kagaku Kogyo Co., Ltd. may be exemplified (all of them are trademarks), though not limited thereto.

For forming the primer coating layer using such a primer paint, the main agent constituting the primer paint and a hardening agent are dissolved in a suitable solvent and are coated on the surface-treated layer on the metal layer, followed by heating at the hardening temperature of the hardening agent used and hardening, if necessary with pressing, to build up the primer coating layer.

The polyolefin modified by carboxyl group or by a derivative thereof to be used as the adhesive resin according to the present invention is a graft-modified polyolefin (in the following, sometimes referred to simply as the polyolefin) in which a not yet modified polyolefin resin is co-polymerized by grafting reaction with preferably at least one polymerizable ethylenically unsaturated carboxylic acid or derivative thereof. As the not yet modified polyolefin resin, there may be exemplified a polyolefin resin, such as polypropylene

and polyethylene. Among them, especially polypropylene is preferable in view of the heat resistance.

As the polypropylene, homopolymers of propylene, copolymers of propylene with, usually, not greater than 20 mole % of an  $\alpha$ -olefin having two or more carbon atoms other than propylene, copolymers of propylene with ethylenically unsaturated carboxylic acids or derivatives thereof and so on may be recited. As the polyethylene, homopolymers of ethylene, copolymers of ethylene with, usually, not greater than 20 mole % of an  $\alpha$ -olefin of 3 or more carbon atoms and copolymers of ethylene with ethylenically unsaturated carboxylic acids or derivatives thereof may be recited. In the case of copolymers, they may either be random copolymers or block-copolymers. For the ethylenically unsaturated carboxylic acids or derivatives thereof mentioned above to be co-polymerized with propylene or with ethylene, the same ones with those which will be described afterward may be used. As the not yet modified polyolefin resin, homopolymers of propylene, copolymer of propylene with  $\alpha$ -olefins, homopolymers of ethylene and copolymers of ethylene with  $\alpha$ -olefins are preferable. They may be used either each alone or in a combination of two or more of them.

In the modified polyolefins, the grafted amount (grafting proportion) of the ethylenically unsaturated carboxylic acid or the derivative thereof with respect to the entire modified polyolefin resin may favorably be 0.001 - 1 % by weight, preferably 0.01 - 0.6 % by weight.

As the ethylenically unsaturated polymerizable carboxylic acid or derivative thereof, which is to be grafted onto the not yet modified polyolefin resin, there may be enumerated, for example, acrylic acid, methacrylic acid, maleic acid, itaconic acid, citraconic acid, mesaconic acid, maleic anhydride, 4-methylcyclohex-4-en-1,2-dicarbocyclic acid anhydride, bicyclo[2.2.2]oct-5-en-2,3-dicarboxylic acid anhydride, 1,2,3,4,5,8,9,10-octahydronaphthalene-2,3-dicarboxylic acid anhydride, 2-octa-1,3-diketospiro[4.4]-non-7-ene, bicyclo[2.2.1]hept-5-en-2,3-dicarboxylic acid anhydride, maleopimamic acid, tetrahydrophtalic acid anhydride,  $\alpha$ -methylbicyclo[2.2.1]hept-5-en-2,3-dicarboxylic acid anhydride,  $\alpha$ -methylnorbornen-5-en-2,3-dicarboxylic acid anhydride and norborn-5-en-2,3-dicarboxylic acid anhydride. Preferably maleic anhydride is used. They may be used either alone or in a combination of two or more of them.

For grafting a graft-monomer selected from the group consisting of ethylenically unsaturated carboxylic acids or derivatives thereof onto the not yet modified polyolefin resin, methods known from the past can be incorporated. For example, a method in which the polyolefin resin is melted and the graft-monomer is added thereto to effect the grafting reaction or a method in which the polyolefin resin is dissolved in a solvent to prepare a solution and the graft-monomer is added thereto to effect the grafting reaction may be recited. In each case, it is preferable to carry out the grafting reaction in the presence of a radical

initiator in order to effect the graft-copolymerization efficiently. The grafting reaction is carried out usually under a condition of 60 - 350 °C. The proportion of radical initiator to be used may usually be in the range of 0.001 - 1 part by weight, per 100 parts by weight of the not yet modified polyolefin resin.

For the radical initiator, organic peroxides may favorably be used and there may be enumerated, for example, benzoyl peroxide, dichlorobenzoyl peroxide, dicumyl peroxide, di-tert-butyl peroxide, 2,5-dimethyl-2,5-di(peroxybenzoate)hexyne-3, 1,4-bis(tert-butylperoxyisopropyl)benzene, lauroyl peroxide, tert-butyl peracetate, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexyne-3, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane, tert-butyl perbenzoate, tert-butylperphenyl acetate, tert-butyl perisobutyrate, tert-butyl per-sec-octoate, tert-butyl perpivalate, cumyl perpivalate and tert-butyl perdiethylacetate. Further, azo compounds, such as azobisisobutyronitrile and dimethylazoisobutyrate, may also be used.

For the radical initiator, at most preferable one for each specific grafting reaction process should be selected, while usually a dialkyl peroxide may be used preferably, such as dicumyl peroxide, di-tert-butyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexyne-3, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane and 1,4-bis(tert-butylperoxyisopropyl)benzene.

The adhesive resin made of the polyolefin modified by carboxyl group or by derivative thereof is

processed into a film or a sheet to form the adhesive resin layer. For such processing, known techniques, such as extrusion molding and inflation molding, can be employed.

The laminate according to the present invention can be produced by effecting a surface treatment on the surface of the metal layer to form a surface-treated layer and laying on the resulting surface-treated layer with a layer of an adhesive resin constituted of the polyolefin modified by carboxyl group or by derivative thereof or, alternatively, coating on the resulting surface-treated layer with a primer paint to form a primer coating layer and laying on the primer coating layer with a layer of an adhesive resin constituted of the polyolefin modified by carboxyl group or by derivative thereof. On forming the primer coating layer, it is preferable to heat and harden it, before laying the adhesive resin layer thereon and heating the laid layer to fusedly bond it, though it is permissible to lay the adhesive resin layer on the primer coating before the hardening thereof and to effect the hardening by making use of the heat upon the fusion bonding.

The laminate according to the present invention produced as above has a lamination structure of metal layer/surface-treated layer/adhesive resin layer or metal layer/surface-treated layer/primer coating layer/adhesive resin layer. In case the primer coating layer is present, the adhesion between the metal layer (surface-treated layer) and the adhesive resin layer is strengthened by the presence of the primer coating

layer. When the non-aqueous electrolyte solution is held in contact with the adhesive resin layer on this side, occurrence of separation at the interface between the metal layer and the primer coating layer caused by the non-aqueous electrolyte solution is prevented by the presence of the surface-treated layer.

On the laminate of the present invention, one or more substrate resin layers constituted of one or more substrate resins, for example, polyolefin resins, such as polyethylene and polypropylene, polyester resins, such as polyethylene terephthalate etc., and others, can be laminated on the side reverse to the metal layer. The lamination structure in this case may be substrate resin layer/metal layer/surface-treated layer/adhesive resin layer or substrate resin layer/metal layer/surface-treated layer/primer coating layer/adhesive resin layer.

The laminate according to the present invention is used as a seal film for sealing the electrolyte solution of primary or secondary batteries or as a protection film for protecting the electrode part of primary or secondary batteries, wherein it is used by holding an organic polar solvent and/or a salt on the side of the adhesive resin layer in contact therewith. By using it, in particular, in a state of contacting with a non-aqueous electrolyte solution containing the organic polar solvent and the salt, it can be used adaptively as a seal film for sealing the electrolyte solution of secondary batteries or as a protection film for protecting the electrode part of secondary

batteries, such as, in particular, non-aqueous electrolyte batteries and solid-state batteries. Here, it is possible to use it as a sealing bag for a battery by folding it so as to face the adhesive resin layer side to each other and heat-sealing it. The adhesive resin layer used according to the present invention is superior in the ability for being subjected to heat-sealing, so that prevention of leakage of non-aqueous electrolyte and long-term use for battery become possible.

For the organic polar solvent, aprotic polar solvents, for example, alkyl carbonates, esters and ketones, may be enumerated. Concrete examples include ethylene carbonate, propylene carbonate, butylene carbonate, dimethyl carbonate, ethyl methyl carbonate, diethyl carbonate,  $\gamma$ -butyrolactone, 1,2-dimethoxyethane, tetrahydrofuran, 2-methyltetrahydrofuran, 1,3-dioxolan, 4-methy-1,3-dioxolan, methyl formate, 4-methyl-1,3-dioxomethyl formate, methyl acetate and methyl propionate.

As the salts, there may be recited alkali metal salts, such as lithium salts, sodium salts and potassium salts. For batteries, lithium salts, such as LiPF<sub>6</sub>, LiBF<sub>4</sub> and Li-imide, are often used.

The non-aqueous electrolyte is one in which an alkali metal salt as given above is dissolved in an amount of 0.5 - 3 mmol in an organic aprotic polar solvent among those of cyclic carbonic acid esters, linear carbonic acid esters and mixtures of them.

The laminate, the seal film for sealing the

electrolyte of battery, the protection film for protecting the electrode part of battery, the seal film for sealing the electrolyte of secondary battery and the protection film for protecting the electrode part of secondary battery according to the present invention can be used over a prolonged period of time without suffering from occurrence of interlayer separation between the metal layer, the surface-treated layer, the primer coating layer and the adhesive resin layer, even when they are used in a state of contacting with the above-mentioned polar solvent and/or the salt, in particular, with the non-aqueous electrolyte which is a mixture of them.

The battery according to the present invention is a battery which has a seal film for sealing the electrolyte of the battery or a protection film for protecting the electrode part of the battery made of the above-mentioned laminate. The battery according to the present invention can be used as a battery in a stable manner over a prolonged period of time, since the film mentioned above does not suffer from interlayer separation and can protect leakage of the non-aqueous electrolyte.

The secondary battery according to the present invention is a secondary battery which has a seal film for sealing the electrolyte of the secondary battery or a protection film for protecting the electrode part of the secondary battery made of the above-mentioned laminate. The secondary battery according to the present invention can be used as a battery in a stable

manner over a prolonged period of time, since the film mentioned above does not suffer from interlayer separation and can protect leakage of the non-aqueous electrolyte.

The flexible package according to the present invention is a flexible packaging material made of the said laminate. The flexible package according to the present invention can be used for, such as a sealing bag for a battery, by folding it so as to face the adhesive resin layer side to each other and heat-sealing it. It can further be used as a sealing bag for, for example, electric and electronic parts, such as condenser etc.

As detailed above, the laminate according to the present invention is superior not only in the adhesive strength between the metal layer and the adhesive resin layer but also in the fastness to organic polar solvents or to salts and does not suffer from occurrence of interlayer separation even in contact with a non-aqueous solvent or the like, since the laminate is made by forming on the metal layer a surface-treated layer and laying thereon the adhesive resin layer under intermediation by a primer coating layer. For this reason, batteries in which the laminate is used as the seal film for sealing the electrolyte of the battery or as the protection film for protecting the electrode part of the battery and secondary batteries in which the laminate is used as the seal film for sealing the electrolyte of the secondary battery or as the protection film for protecting the electrode part of the secondary battery

can be used in a stable manner over a prolonged period of time.

#### THE BEST MODE FOR EMBODYING THE INVENTION

Below, the present invention will be described by way of Examples of the present invention, in which % values are given on the weight basis.

##### EXAMPLE 1

###### « Preparation of Test Specimen»

An aluminum plate (JIS-H419, 140×60×0.2 mm) was immersed in a chemical treatment liquor of phosphoric acid/chromic acid as a surface-treating agent in pure water (a liquid in which a trivalent chromium concentration was adjusted at 0.24 %) and then washed with water, followed by drying in an air oven of 100 °C for 20 minutes, whereby a treated aluminum plate specimen A was prepared.

A layer of a maleic anhydride-modified polypropylene resin (MFR = 3g/10 min., grafted amount of maleic anhydride = 0.15 %, film thickness = 50 μm) as the adhesive resin was laid over the treated aluminum specimen A and the resulting laminate was press-bonded [preheating: 180 °C × 3 min., pressing: 180 °C × 1 min. under a pressure of 0.59 MPa (6 kg/cm<sup>2</sup>), cooling: 15 °C × 3 min. under a pressure of 0.49 MPa (5 kg/cm<sup>2</sup>)]. Assessment of the adhesive strength onto the aluminum specimen gave a value of 8.0 N/15 mm.

The condition of peeling:

15 mm width, 180° peeling, 50 mm/min. (i.e. a laminate having a width of 15 mm was tested by peeling off in the direction of 180° at a peeling velocity of 15 mm/min.; the same applies to all appearing afterwards).

« Durability Immersion Tast »

An electrolyte in which the proportions of ethylene carbonate, methyl ethyl carbonate and LiPF<sub>6</sub> are adjusted at 30 %, 55 % and 15 %, respectively, was poured into a cylindrical stainless steel kettle of a capacity od one liter, whereupon the specimen described above was immersed in the electrolyte. Then the kettle was sealed completely with a lid so as not to permit leakage of the liquid and was stood still under a state heated at 80 °C in a constant temperature bath for 10 days. Then, the stainless steel kettle was opened after it was cooled to room temperature and the specimen was taken out, followed by sufficient washing off of the electrolyte. Assessment of the adhesive strength onto the aluminum specimen gave a value of 4.5 N/15 mm.

The condition of peeling:

180° peeling, 50 mm/min.

EXAMPLE 2

In the procedures of EXAMPLE 1, a treated aluminum plate specimen B was prepared by the surface-treatment and subsequent application of DUFLON K300 (trademark) of Nippon Paint Mfg. Co., Ltd. as the primer coating based on epoxy with a pretreatment performed by heating at 170 °C × 15 min. in an oven. Assessment of the adhesive strength in the same manner

as in Example 1 gave a result of impossible peeling off both before and after the immersion (the adhesive resin destroyed the substrate resin).

EXAMPLE 3

Test was carried out in the same manner as in EXAMPLE 2, except that ORGAPLASUF 30NC (trademark) of Nippon Paint Mfg. Co., Ltd. was used as the primer coating based on epoxy. The results are recited in Table 1.

EXAMPLE 4

Test was carried out in the same manner as in EXAMPLE 2, except that EPOMATE B002/EPIKOTE 828 (trademark) of Yuka-Shell K.K. was used as the primer coating based on epoxy and the pretreatment was carried out by heating at 180 °C × 10 min. The results are recited in Table 1.

EXAMPLE 5

Test was carried out in the same manner as in EXAMPLE 2, except that ETON 2100E (trademark) of Kawakami Paint Mfg. Co., Ltd. was used as the primer coating based on epoxy and the pretreatment was carried out by heating at 120 °C × 20 min. The results are recited in Table 1.

EXAMPLE 6

Test was carried out in the same manner as in EXAMPLE 2, except that maleic anhydride-modified polypropylene (MFR = 7 g/10 min., grafted amount of maleic anhydride = 0.15 %) was used for the adhesive resin film. The results are recited in Table 1.

EXAMPLE 7

Test was carried out in the same manner as in EXAMPLE 2, except that maleic anhydride-modified polyethylene (MFR = 1.0 g/10 min., grafted amount of maleic anhydride = 0.11 %) was used for the adhesive resin film. The results are recited in Table 1.

EXAMPLE 8

Test was carried out in the same manner as in EXAMPLE 2, except that maleic anhydride-modified polyethylene (MFR = 2.9 g/10 min., grafted amount of maleic anhydride = 0.18 %) was used for the adhesive resin film. The results are recited in Table 1.

EXAMPLE 9

Test was carried out in the same manner as in EXAMPLE 2, except that CORONATE L/NIPPORAN 1100 (trademark) of Nippon Polyurethane Industry Co., Ltd. was used as the primer paint based on urethane and the pretreatment was carried out by heating at 80 °C × 5 minutes. The results are recited in Table 1.

EXAMPLE 10

Test was carried out in the same manner as in EXAMPLE 2, except that A-310/A-3 (trademark) of Takeda Chemical Ind., Ltd. was used as the primer paint based on urethane and the pretreatment was carried out by heating at 80 °C × 5 minutes. The results are recited in Table 1.

EXAMPLE 11

Test was carried out in the same manner as in EXAMPLE 2, except that P-1000 (trademark) of Nippon Shokubai Kagaku Kogyo Co., Ltd. was used as the primer paint based on imine and the pretreatment was carried

out by heating at 80 °C × 10 minutes. The results are recited in Table 1.

COMPARATIVE EXAMPLE 1

An aluminum plate was immersed in a toluene solution and was subjected to an ultrasonic cleaning for 20 minutes. In the same manner as in EXAMPLE 2, the primer coating layer and the adhesive resin layer were laminated on the cleaned aluminum plate without forming a surface-treated layer thereon, whereupon the test was carried out. The results are recited in Table 1.

COMPARATIVE EXAMPLE 2

An aluminum plate was immersed in a toluene solution and was subjected to an ultrasonic cleaning for 20 minutes. In the same manner as in EXAMPLE 3, the primer coating layer and the adhesive resin layer were laminated on the cleaned aluminum plate without forming a surface-treated layer thereon, whereupon the test was carried out. The results are recited in Table 1.

COMPARATIVE EXAMPLE 3

An aluminum plate was immersed in a toluene solution and was subjected to an ultrasonic cleaning for 20 minutes. In the same manner as in EXAMPLE 4, the primer coating layer and the adhesive resin layer were laminated on the cleaned aluminum plate without forming a surface-treated layer thereon, whereupon the test was carried out. The results are recited in Table 1.

COMPARATIVE EXAMPLE 4

An aluminum plate was immersed in a toluene solution and was subjected to an ultrasonic cleaning for 20 minutes. In the same manner as in EXAMPLE 5, the

primer coating layer and the adhesive resin layer were laminated on the cleaned aluminum plate without forming a surface-treated layer thereon, whereupon the test was carried out. The results are recited in Table 1.

COMPARATIVE EXAMPLE 5

Test was carried out in the same manner as in Example 2 except that a film of the adhesive resin was laid directly on the aluminum plate which had been subjected to a surface treatment in the same manner as in Example 2 without forming thereon the primer coating layer based on epoxy and the laminate was press-bonded [preheating: 180 °C × 3 minutes, pressing: 180 °C × 1 minute under a pressure of 0.59 MPa (6 kg/cm<sup>2</sup>), cooling: 15 °C × 3 minutes under a pressure of 0.49 MPa (5 kg/cm<sup>2</sup>)]. The results are recited in Table 1.

Table 1

	Adhesive Strength (N/15 mm)	
	Before Immersion	After Imm.
<u>EXAMPLE</u>		
1	8.0	4.5
2	Unpeelable (substr. destr. *)	Unpeelable
3	Unpeelable (substr. destr.)	Unpeelable
4	Unpeelable (substr. destr.)	3.3
5	Unpeelable (substr. destr.)	6.8
6	Unpeelable (substr. destr.)	Unpeelable
7	11.2	10.5
8	11	8.8
9	10.8	5.6
10	9.6	4.5
11	5.8	1.2
<u>COMPAR. EXAMPLE</u>		
1	Unpeelable (substr. destr.)	0 (peeled)
2	Unpeelable (substr. destr.)	0 (peeled)
3	Unpeelable (substr. destr.)	0 (peeled)
4	Unpeelable (substr. destr.)	0 (peeled)
5	Unpeelable (substr. destr.)	0 (peeled)

Note: \* substrate destroyed

#### INDUSTRIAL APPLICABILITY

The laminate according to the present invention is superior not only in the adhesive strength between the metal layer and the adhesive resin layer but also

in the fastness to organic polar solvents or to salts and does not suffer from occurrence of interlayer separation upon contact with non-aqueous electrolyte and the like, so that it can be used adaptively as protection film for protecting the electrode part of primary and secondary batteries or as protecting film for protecting the electrode part of secondary batteries, whereby it is made possible to obtain primary or secondary batteries which can be used in a stable manner for prolonged period of time.

CLAIMS

1. A laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising
  - a metal layer,
  - a surface-treated layer formed over the surface of the metal layer and
  - a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.
2. A laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising
  - a metal layer,
  - a surface-treated layer formed over the surface of the metal layer,
  - a layer of a primer coating formed over the said surface-treated layer and
  - a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.
3. A laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, comprising
  - a metal layer,
  - a surface-treated layer formed over the surface of the metal layer and
  - a layer of an adhesive resin of a polyolefin

modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.

4. A laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer,

a layer of a primer coating formed over the said surface-treated layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.

5. The laminate as claimed in any one of claims 1 to 4, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

6. The laminate as claimed in any one of claims 1 to 5, wherein the surface-treated layer is constituted of a chemically treated layer.

7. The laminate as claimed in any one of claims 1 to 6, wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

8. The laminate as claimed in any one of claims 1 to 7, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

9. The laminate as claimed in claim 8, wherein the graft-modified polyolefin resin is a graft-modified polyethylene resin or a graft-modified polypropylene resin.

10. A process for producing a laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising

forming a surface-treated layer over the surface of a metal layer and

laminating on the surface-treated layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

11. A process for producing a laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising

forming a surface-treated layer over the surface of a metal layer,

forming a primer coating layer over the said surface-treated layer and

laminating on the primer coating layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

12. A process for producing a laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, comprising

forming a surface-treated layer over the surface of a metal layer and

laminating on the surface-treated layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

13. A process for producing a laminate to be served as seal film for sealing the electrolyte of secondary battery or as protection film for protecting the electrode part of secondary battery, comprising

forming a surface-treated layer over the surface of a metal layer,

forming a primer coating layer over the said surface-treated layer and

laminating on the primer coating layer a layer of an adhesive resin of a polyolefin resin modified by carboxyl group or a group derived therefrom.

14. A seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in any one of claims 1, 2 and 5 to 9.

15. A battery comprising a seal film for sealing the electrolyte of battery or a protection film for protecting the electrode part of battery, which film is made from the laminate as claimed in any one of claims 1, 2 and 5 to 9.

16. A seal film for sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in any one of claims 3 to 9.

17. A secondary battery comprising a seal film for

sealing the electrolyte of secondary battery or a protection film for protecting the electrode part of secondary battery, which film is made from the laminate as claimed in any one of claims 3 to 9.

18. A flexible package made from a laminate comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said surface-treated layer.

19. A flexible package made from a laminate comprising

a metal layer,

a surface-treated layer formed over the surface of the metal layer,

a layer of a primer coating formed over the said surface-treated layer and

a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom, formed over the said primer coating layer.

20. The flexible package as claimed in claim 18 or 19, wherein the metal layer is constituted of one or more metals selected from the group consisting of aluminum, nickel, copper, iron and alloys of them.

21. The flexible package as claimed in any one of claims 18 to 20, wherein the surface-treated layer is constituted of a chemically treated layer.

22. The flexible package as claimed in any one of

claims 18 to 21, wherein the primer coating layer is that of thermosetting type based on epoxy, urethane, epoxy-urethane, imine, titanate, polyester or silane.

23. The flexible package as claimed in any one of claims 18 to 22, wherein the adhesive resin is a graft-modified polyolefin resin obtained by graft-copolymerizing a polyolefin resin with an ethylenically unsaturated carboxylic acid.

24. The flexible package as claimed in claim 23, wherein the graft-modified polyolefin resin is a graft-modified polyethylene resin or a graft-modified polypropylene resin.

## ABSTRACT

A laminate to be served as seal film for sealing the electrolyte of battery or as protection film for protecting the electrode part of battery, comprising a metal layer, a surface-treated layer formed over the surface of the metal layer and a layer of an adhesive resin of a polyolefin modified by carboxyl group or a group derived therefrom formed over the said surface-treated layer, which laminate is superior in the adhesive strength between the metal layer and the adhesive resin layer and in the fastness to organic polar solvents or to salts and, therefore, does not suffer from occurrence of interlayer separation on contacting with a non-aqueous solvent and, hence, can favorably be used in particular as seal film for sealing the electrolyte of secondary battery.

# BIRCH, STEWART, KOLASCH & BIRCH, LLP

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# LAMINATE, PROCESS FOR PRODUCING IT AND USE THEREOF

the specification of which is attached hereto. If not attached hereto,

the specification was filed on \_\_\_\_\_ as  
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I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I do not know and do not believe the same was ever known or used in the United States of America before my or our invention thereof, or patented or described in any printed publication in any country before my or our invention thereof or more than one year prior to this application, that the same was not in public use or on sale in the United States of America more than one year prior to this application, that the invention has not been patented or made the subject of an inventor's certificate issued before the date of this application in any country foreign to the United States of America on an application filed by me or my legal representatives or assigns more than twelve months (six months for designs) prior to this application, and that no application for patent or inventor's certificate on this invention has been filed in any country foreign to the United States of America prior to this application by me or my legal representatives or assigns, except as follows.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

**Insert Priority  
Information:  
(if appropriate)**

Prior Foreign Application(s)			Priority	Claimed
242188/1999 (Number)	Japan (Country)	08/27/1999 (Month/Day/Year Filed)	<input checked="" type="checkbox"/> Yes	<input type="checkbox"/> No
<hr/> (Number)	<hr/> (Country)	<hr/> (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
<hr/> (Number)	<hr/> (Country)	<hr/> (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
<hr/> (Number)	<hr/> (Country)	<hr/> (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No
<hr/> (Number)	<hr/> (Country)	<hr/> (Month/Day/Year Filed)	<input type="checkbox"/> Yes	<input type="checkbox"/> No

I hereby claim the benefit under Title 35, United States Code, §119(e) of any United States provisional application(s) listed below.

**Insert Provisional  
Application(s):  
(if any)**

---

(Application Number) \_\_\_\_\_ (Filing Date) \_\_\_\_\_

All Foreign Applications, if any, for any Patent or Inventor's Certificate Filed More Than 12 Months (6 Months for Designs) Prior To The Filing Date of This Application:

**Insert Requested  
Information:  
(if appropriate)**

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

**Insert Prior U.S.  
Application(s):  
(if any)**

(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)
(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)

I hereby appoint the following attorneys to prosecute this application and/or an international application based on this application and to transact all business in the Patent and Trademark Office connected therewith and in connection with the resulting patent based on instructions received from the entity who first sent the application papers to the attorneys identified below, unless the inventor(s) or assignee provides said attorneys with a written notice to the contrary:

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PLEASE NOTE:  
YOU MUST  
COMPLETE THE  
FOLLOWING:

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full Name of First or Sole Inventor:

Insert Name of Inventor  
Insert Date This Document is Signed

Insert Residence  
Insert Citizenship

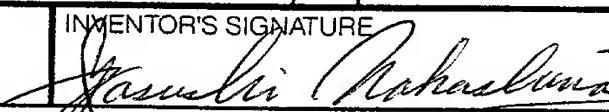
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GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
Residence (City, State & Country)		CITIZENSHIP	
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GIVEN NAME	FAMILY NAME	INVENTOR'S SIGNATURE	DATE*
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\* DATE OF SIGNATURE